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(54) PHOTO-REFRACTIVE MATERIAL COMPOSITION

(57) Abstract:

PROBLEM TO BE SOLVED: To obtain a stably usable practical polymer-base photo-refractive material having high-speed responsiveness by regulating hole mobility under the conditions of a specified applied electric field and a specified temp. to above a specified value.

SOLUTION: In a photo-refractive material compsn. contg. a hole transferring polymer contg. a triphenylamine deriv. and nonlinear optical molecules, hole mobility under the conditions of 1×107 V/m applied electric field and 25°C is regulated to $>1 \times 10-6$ cm²/V.sec. The triphenylamine deriv. may be introduced into the principal chain of the hole transferring polymer or into the polymer as a side chain by covalent bond or may be dispersed in the polymer but it is preferable that the deriv. is covalently bonded as a side chain. The polymer may be a homo- or copolymer.

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CLAIMS

[Claim(s)]

[Claim 1] The photorefractive ingredient constituent with which Hall mobility under impression electric-field 1x107 V/m and conditions with a temperature of 25 degrees C is characterized by the bigger thing than 1x10-6cm2 / V-sec in the photorefractive ingredient constituent containing the hole transportability polymer containing a triphenylamine derivative, and a nonlinear optics molecule.

[Claim 2] The photorefractive ingredient constituent according to claim 1 with which a hole transportability polymer is characterized by having a triphenylamine derivative in a side chain.

[Claim 3] The photorefractive ingredient constituent according to claim 2 characterized by including the copolymer of the acrylate which has N, N'-diphenyl-N, N'-screw (4-methylphenyl)-[1 and 1'-biphenyl]-4, and 4'-diamine in a side chain as a hole transportability polymer, and n-butyl acrylate.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[Field of the Invention] When this invention carries out an optical exposure, it is used for the various components of the medium of optical recording, especially holographic record, or optical information processing about the photorefractive ingredient which produces the refractive-index distribution according to the exposure pattern. [0002]

[Description of the Prior Art] The phenomenon in which the refractive-index pattern of the matter changes according to a photorefractive effect, i.e., the irradiated pattern on the strength [optical], is P.Guenter. And you could set to inorganic crystals, such as lithium niobate (LiNbO3), and barium titanate (BaTiO3), a silicic acid bismuth (Bi12SiO20), and it was known as stated to "Photorefractive Materials and Their Applications I&II" (Springer-Verlag, 1988) edited by J.P.Huignard. the trap of generating of the charge according [the process of the refractive-index change by the optical exposure in a photorefractive ingredient] to (1) light exposure, diffusion of (2) charges or the drift by electric field, and (3) charges, formation of the electric field by (4) space charge, formation of the refractive-index distribution by (5) electro-optical effects, and ** -- it thinks like. Therefore, the trapping of (1) photoconductivity (charge generating and charge transportation) and (2) charges and the (3) primary electro-optical effect (Pockels effect) are required of a photorefractive ingredient. Inorganic crystals, such as lithium

niobate, barium titanate, and a silicic acid bismuth, are ingredients which fulfill these conditions. However, since sensibility and a speed of response have the relation of a trade-off as for these inorganic crystals, raising both both has the fault that it is difficult and inferior to a moldability and workability.

[0003] Then, the photorefractive ingredient of a polymer system is proposed as what conquers the fault of these inorganic crystals. or it makes JP,6-55901,B distribute a carrier transportation molecule and a nonlinear optics molecule in a polymer binder -- or the molecule concerned -- covalent bond -- the inside of the principal chain of a polymer -- or the photorefractive ingredient introduced as a side chain is indicated. moreover --JP,5-265062,A -- the outside of a carrier transportation molecule and a nonlinear optics molecule -- further -- a carrier trap -- the inside of a principal chain -- or the photorefractive polymer introduced as a side chain is also indicated. It is equal to an inorganic crystal also about the practical use property of such a polymer system photorefractive ingredient, or the value beyond it is acquired. To the Pori (4-n-butoxy phenyl) ethyl silane of a photoconductivity, as a nonlinear optics molecule (E)-betanitroglycerine-(Z)-beta-methyl-3-fluoro-4-N and N-diethylamino styrene Moreover, it sets to degenerate four wave mixing which used the polymer system which added fullerene C60 as a charge generating agent. Impression electric-field 1.14x107 V/m, Record light on-the-strength 1 W/cm2 The value of a high speed called 39msec(s) is acquired as a time constant of a diffraction-efficiency standup under conditions (S. M.Silence et al., J.Opt.Soc.Am.B, Vol.10, No.12, 2306, and 1993). moreover, the polyvinyl carbazole of hole transportability -- as a nonlinear optics molecule -- a 2 and 5dimethyl-4-(p-nitro phenylazo) anisole -- as a charge generating agent -- trinitro full -me -- in the polymer system which added N-ethyl carbazole as a plasticizer for lowering glass transition temperature for non, about 90% of diffraction efficiency is acquired (KMeerholz etal., Nature, Vol.371, 497, and 1994).

[Problem(s) to be Solved by the Invention] Also when it is required that a speed of response should be quick when using a photorefractive ingredient for a display and optical signal processing of an animation, and using as material for optical recording, in order to gather a transfer rate, high-speed responsibility is required. For example, when performing a movie display, the speed of response of the minimum video rate, i.e., the speed of response of 30msec(s), is required. on the other hand, as a typical value of the speed of response of an actual polymer system photorefractive ingredient With the ingredient of a polyvinyl-carbazole system, the record light reinforcement of 1W/cm under the conditions of 2 and impression electric-field 4x107 V/m the value of about 100 msec(s) is acquired as a time constant of the diffraction-efficiency standup in 4 lightwave mixing (M. C.J.M.Donckers, et al., Optic Letters, Vol.18, No.13, 1044, 1993) -- it is . moreover, with the ingredient used as the base, a Pori (4-n-butoxy phenyl) ethyl silane as mentioned above Although the value of high-speed 39msec(s) is acquired more as a time constant of the diffraction-efficiency standup in 4 light-wave mixing under the conditions of record light on-the-strength 1 W/cm2 and impression electric-field 1.14x107 V/m Since it is unstable, and it is easy to decompose to light and the ingredient of this polysilane system has the fault that a mechanical strength is also weak, it must be told to practicality that it is scarce. As mentioned above, about a polymer system photorefractive ingredient, the charge of real material in which a high-speed response is

possible is not yet found. Therefore, the purpose of this invention has high-speed responsibility, and is to offer the practical polymer system photorefractive ingredient which can be used for stability.

[0005]

[Means for Solving the Problem] The above-mentioned technical problem is solvable in the photorefractive ingredient constituent containing the hole transportability polymer containing a triphenylamine derivative, and a nonlinear optics molecule by considering as the photorefractive ingredient constituent with which Hall mobility under impression electric-field 1x107 V/m and conditions with a temperature of 25 degrees C is characterized by the bigger thing than 1x10-6cm2 / V-sec.

[0006] That is, this invention offers the following.

- [1] The photorefractive ingredient constituent with which Hall mobility under impression electric-field 1x107 V/m and conditions with a temperature of 25 degrees C is characterized by the bigger thing than 1x10-6cm2 / V-sec in the photorefractive ingredient constituent containing the hole transportability polymer containing a triphenylamine derivative, and a nonlinear optics molecule.
- [2] The photorefractive ingredient constituent of the aforementioned [1] publication with which a hole transportability polymer is characterized by having a triphenylamine derivative in a side chain.
- [3] as a hole transportability polymer -- N and N' diphenyl-N and N' screw (4-methylphenyl)-[1 and 1'-biphenyl]-4 and 4' -- the photorefractive ingredient constituent of the aforementioned [2] publication characterized by including the copolymer of the acrylate which has diamine in a side chain, and n-butyl acrylate.

This invention is explained below at a detail.

[0007] The triphenylamine derivative which has played a role of a hole transportation agent in the constituent of this invention gives the photorefractive constituent which has bigger Hall mobility than 1x10-6cm2 / V-sec under impression electric-field 1x107 V/m and conditions with a temperature of 25 degrees C. Incidentally as typical mobility of the conventional polymer system photorefractive constituent In the system which added non the polyvinyl carbazole which is a hole transportability polymer -- as a nonlinear optics molecule -- the De Dis Perth red 1 -- as a charge generating agent -- 2, 4, and 7-trinitro-9-full -- me -- The value of 7x10-6cm2 / V-sec is acquired at the room temperature at the time of impression electric-field 108 V/m (S. B.Kippelen and N.Peyghambarian, Optics Letters, Vol.19, No.1, 68, 1994). If it converts when it carries out based on this value and impression electric field are 107 V/m, mobility will become below 1x10-7cm2 / V-sec. It becomes possible to acquire high-speed responsibility by using the ingredient which has big mobility compared with the conventional polymer system ingredient like the constituent of this invention.

[0008] the hole transportability polymer containing a triphenylamine derivative -- covalent bond -- a triphenylamine derivative -- the inside of a principal chain -- or although it can also introduce as a side chain and can also be made to distribute in a polymer, it is desirable to carry out covalent bond as a side chain. The constituent of this invention contains about 50% of the weight or more of a triphenylamine derivative preferably about 20% of the weight or more. Moreover, a homopolymer or a copolymer is sufficient as the polymer in the constituent of this invention. This polymer has one or less [100cm -] optical absorption coefficient preferably in bigger number average

molecular weight than about 5000 and the wavelength to be used.
[0009] The triphenylamine derivative which can be used for the constituent of this invention is the compound of the following structures.
[Formula 1]

$$R_1 \bigoplus R_1$$
 $R_1 \bigoplus R_1$
 $R_1 \bigoplus R_2$

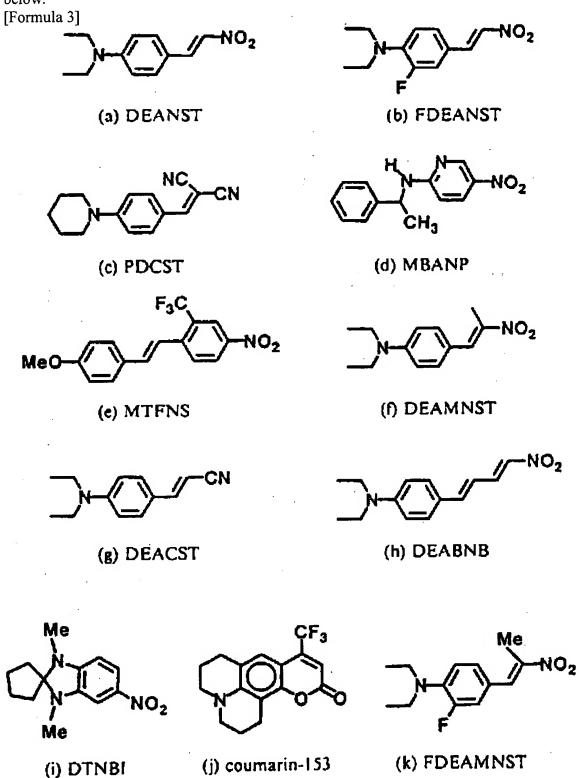
[R1 is H, m-Me (= methyl group), p-Me, etc. among a formula, and R2 is H, Me, 2 (p-Me-C six H4) N, etc.]

$$\begin{array}{c|c} & & & & & & & \\ R_3 & & & & & & \\ \hline R_4 & & & & & & \\ \hline \end{array}$$

[R3 is H, p-Me, p-Et, etc. among a formula, R4 is H, m-Me, p-Me, p-Et, o-Me, etc., and R5 is H, Me, etc.]

[0010] the nonlinear optics molecule in the constituent of this invention -- covalent bond -- the inside of a principal chain -- or it can also introduce into a polymer as a side chain, and can also be made to distribute in a polymer the case where it is used for an application by which a secondary eternal nonlinear optics response (primary electro-optics response) is needed for this constituent -- a nonlinear optics molecule -- the inside of a principal chain -- or introducing as a side chain is desirable, and when used for an application for which high-speed responsibility is needed, it is desirable to distribute a nonlinear optics molecule in a polymer. The constituent of this invention contains 5 - 50% of the weight of a nonlinear optics molecule preferably about 1% of the weight or more. What has secondary nonlinear optics nature as a nonlinear optics molecule in the constituent of this invention is usable, and what was indicated by "Nonlinear OpticalProperties of Organic Molecules and Crystals" (Academic Press, 1987) edited by

D.S.Chemla and J.Zyss is known by this. The example of a typical compound is shown below.



[0011] In the constituent of this invention, the ingredient of the principal chain which forms a polymer with a triphenylamine derivative and/or a nonlinear optics molecule has

the usable ingredient good [polyacrylate, polyester, polystyrene, a polyamide, polyurethane, and] and known [especially if there is no absorption in the wavelength to be used, is not limited, and]. Moreover, it is not limited especially if the comonomer in the case of forming a copolymer does not have absorption in the wavelength to be used, either, and the monomer of the above-mentioned polymer etc. is usable. Moreover, when charge generating with the constituent of this invention sufficient on use light source wavelength is not acquired, a charge generating agent may be added. although especially the charge generating agent to add is not what is limited -- C 60, 2, and 4 and 7-trinitro-9-full -- me -- non etc. is usable. The addition of these charge generating agents is 3 or less % of the weight preferably.

[0012] Although the trap function of a charge is obtained even if especially the constituent of this invention does not add a charge trap agent, it is possible by adding a trap agent if needed to reinforce a trap function. What is necessary is just to add the molecule which has oxidation potential lower than the triphenylamine derivative which is a hole transportation agent as a trap agent, since the charge conveyed in the constituent of this invention is a hole. In adding a trap agent to this constituent, it adds 0-about 10%. [0013]

[Function] If light with the fixed pattern on the strength in the constituent of this invention is irradiated, a charge (a hole and electron) will be generated by the nonlinear optics molecule or the added charge generating agent. Among these, a hole carries out hopping conduction of between triphenylamine derivatives, is captured by the charge trap in the nearby **** exposure section, and produces the bias of a charge by this. For this reason, space electric field are formed and the refractive-index pattern corresponding to an optical exposure pattern is formed of the primary electro-optical effect (secondary nonlinear optical effect). Here, a speed of response until the formation rate of space electric field is quick since the triphenylamine derivative used for the constituent of this invention has large mobility, and it results in refractive-index pattern formation becomes quick.

[0014]

[Example] Although an example explains this invention concretely below, thereby, this invention is not limited at all.

[0015] Composition of the copolymer of [Example] 1 N, N'-diphenyl-N, N'-screw (4-methylphenyl)-[1 and 1'-biphenyl]-4, and the acrylate which has 4'-diamine (it omits Following TPD) in a side chain and n-butyl acrylate. The above-mentioned copolymer was compounded according to the following five processes.

(Process 1)

Composition of TPD.

N, N'-diphenyl-[1 and 1'-biphenyl]-4, and 4' - diamine 20g (59.5mmol), 4-iodine toluene 311g (1430mmol), 36.5g (246mmol) of anhydrous potassium carbonate, 30.4g (160mmol) of cuprous iodides, and 18-crown -6 5.55g (21mmol) mixture was heated at 200 degrees C under churning for 24 hours. Toluene 200ml was added and filtered after cooling. After distilling off a solvent and unreacted 4-iodine toluene, it refined by column chromatography (silica gel, a toluene:hexane = 50:50), and specified substance TPD20g of the following structure was obtained. 81% of yield. [Formula 4]

[0016] (Process 2)

N-(4-formyl phenyl)-N'-phenyl-N, N'-screw (4-methylphenyl)-[1 and 1'-biphenyl]-4, composition of 4'-diamine (it abbreviates to TPD-CHO below).

Under nitrogen-gas-atmosphere mind, at 0 degree C, 5.94g (38.7mmol) of phosphoryl chlorides was dropped at desiccation dimethylformamide (DMF) 10ml, and it agitated for 2.5 hours. Then, TPD20g (38.7mmol) obtained at the process 1 and desiccation DMF50ml were added, and it agitated at 80 degrees C for 6 hours. 15g of sodium acetate which put in ice was filled with the reactant after cooling. The solid content which deposited was dried after 2 times washing with water, unreacted TPD and an unreacted JIFORUMIRU ghost were refined by column chromatography (silica gel, a toluene:hexane = 67:33->100:0), and specified substance TPD-CHO9.18g of the following structure was obtained as yellow powder. 45% of yield. [Formula 5]

[0017] (Process 3)

N-(4-hydroxy methylphenyl)-N'-phenyl-N, N'-screw (4-methylphenyl)-[1 and 1'-biphenyl]-4, composition of 4'-diamine (it abbreviates to TPD-CH2 OH below).

1.02g (26.9mmol) of sodium borohydrides was added after dissolving TPD-CHO11.7g (21.5mmol) obtained at the process 2 in benzene / 160ml of ethanol (1:1) mixed solvents under nitrogen-gas-atmosphere mind, and it agitated at the temperature of arbitration for 2 hours. After distilling off the solvent and washing with water, the rough product was dissolved in benzene. The hexane was filled with this and specified substance TPD-CH2 OH9.97g of the following structure was obtained as yellow powder. 85% of yield. [Formula 6]

[0018] (Process 4)

N-(4-acryloyloxy methylphenyl)-N'-phenyl-N, N'-screw (4-methylphenyl)-[1 and 1'-biphenyl]-4, composition of 4'-diamine (it omits Following TPDac).

TPD-CH2 OH8.82g (16.2mmol) obtained by desiccation DMF40ml at the process 3 under nitrogen-gas-atmosphere mind was dissolved. This solution was cooled at 0 degree C, the solution which dissolved chlorination acryloyl 1.83g (20.2mmol) distilled to this in tetrahydrofuran (THF) 15ml was dropped, and it agitated for 2 hours. Churning was performed for further 10 hours, after returning to a room temperature. THF was distilled off after filtering and removing the salt which deposited. The obtained solid-state was refined by column chromatography (silica gel, a toluene:hexane = 67:33->100:0) after washing with water, and specified substance TPDac4.94g of the following structure was obtained. 51.1% of yield.

[0019] (Process 5)

After putting TPDac2.0g (3.325mmol) obtained at process 4, n-butyl acrylate [which was distilled]g [0.67] (5.227mmol) and 2, and 2'-azobis (isobutyronitrile) 14mg (0.0853mmol) into polymer-ized polymerization tubing and performing a nitrogen purge, distilled benzene 8.55ml was put in and heat-sealed, and the polymerization was carried out at 60 degrees C for 24 hours. after [opening] acetone 100ml -- it reprecipitated by having supplied to inside and filtration recovered the polymer. By performing throwing in the collected benzene solution of a polymer in an acetone, and making it reprecipitate further 2 to 3 times, the after [recovery] vacuum drying was refined and carried out, and 2.52g of specified substance of the following structure was obtained as white powder. 94% of yield.

[Formula 8]

$$\begin{array}{c}
\left(CH_{2}-CH\right)_{n} \\
\left(CH_{2}-CH\right)_{n} \\
C_{4}H_{7}
\end{array}$$

$$\begin{array}{c}
C_{4}H_{7} \\
C_{H_{3}}
\end{array}$$

For number average molecular weight, 15000 and weight average molecular weight were [the obtained copolymer / 87.0 degrees C and the maximum absorption wavelength of 95000 and glass transition temperature] 313nm and 354nm, including TPD 64.6% of the weight.

[0020] measurement of the mobility of the photorefractive constituent which used as the base the copolymer obtained in the [example 2] example 1 -- Time-of-flight -- it carried out by law. The component for measurement is the following, and was made and produced. First, vacuum deposition of the aluminum was carried out on the glass substrate (cover glass), and the spin coat of the titanylphthalocyanine distribution solution was carried out on it. Then, the copolymer 90 weight section and the nonlinear optics molecule 4 which were obtained in the example 1 - (N and N-diethylamino. -(beta)- after applying the sample solution (0.25g/(ml)) which dissolved the nitro styrene (DEANST) 10 weight section in 1,2-dichloroethane on the titanylphthalocyanine film by the bar coating machine and making it dry at 90 degrees C for 1 hour, the translucent golden electrode was attached with vacuum deposition) The thickness of the sample layer in this component was 10 micrometers.

[0021] Measurement of the mobility by the Time-of-flight method was performed at the room temperature (25 degrees C) using the system of measurement shown in <u>drawing 1</u> R> 1. Electric field were impressed between the two electrodes of a component, and the

monitor of the photocurrent when irradiating with a xenon lamp from a translucent golden electrode side was carried out with the digitizing oscilloscope. Although a typical photocurrent wave is shown in <u>drawing 2</u>, it is a transition duration tT about the time amount from the point in time of an optical exposure to the part of a shoulder. It carried out and asked for mobility mu by the following formulas.

Thickness V of mu=L 2 / tT, and VL:sample layer: Impression electric field, consequently mobility as shown in drawing 3 were obtained considering impression electric field as a parameter. Here, mobility in case impression electric field are 3x107 V/m is 2x10-5 V/cm2 and sec, and the value of 8x10-6 V/cm2 and sec was acquired as mobility in case impression electric field are 1x107 V/m as an extrapolation number. [0022] The joint gain coefficient by the 2 light-wave alligation of the photorefractive constituent which used as the base the copolymer obtained in the [example 3] example 1 was measured. The photorefractive element for measurement is the following, and was made and produced. First, the copolymer 84.9 weight section, 4 which were obtained in the example 1 - (N and N-diethylamino. -(beta)- the nitro styrene (DEANST) 14.9 weight section and C600.2 weight section were dissolved in 1,2-dichloroethane (0.25g/(ml)), and the sample solution was produced) Next, after carrying out the cast of the sample solution on the ITO electrode and being air-dry at a room temperature for 3 hours, the vacuum drying was performed at the room temperature for 12 hours. Then, the substrate was warmed at 60 degrees C, on both sides of the 100-micrometer Teflon spacer, lamination and weight were carried and two substrates were left at the room temperature for 12 hours. The sample with a thickness of 100 micrometers was obtained by this approach. Measurement of the joint gain coefficient by 2 light-wave alligation was performed using the optical system shown in drawing 4.

[0023] The 633nm beam by which outgoing radiation is carried out from helium-Ne laser is divided into two (reinforcement is 250 mW/cm2 and is equal), and it was made to cross within a sample by the beam splitter (a crossed axes angle is 21.6 degrees). The angle of incidence to the sample of each beam made the beam 1 38.2 degrees, and made the beam 2 60.8 degrees. The photodiode was used for measurement of transmitted beam reinforcement. The impression electric field to a sample are made into 5x107 V/m or 7x107 V/m here. turn on a beam 2 (pump light) after 2 seconds, and the transmitted light reinforcement of the beam 1 when turning off after 9 seconds is measured. Then, the beam 1 (probe light) was turned on after 2 seconds, the transmitted light reinforcement of the beam 2 when turning off after 9 seconds was measured, and unsymmetrical energy transfer as shown in drawing 5 was obtained (when field strength is 5x107 V/m, both measurement results are drawn on drawing in piles). It asked for the joint gain coefficient gamma by the following formulas using this measured value.

gamma= (cos theta/d) [ln(gamma0 beta)-ln (beta+1-gamma0)]

beta: Intensity ratio gamma0 =P/P0 (P0: transmitted-light reinforcement in case there is no pump light, P: transmitted light reinforcement in case there is pump light) of a beam theta: -- incident angle d: of a beam -- the thickness [0024] of a sample Consequently, it was gamma=40cm-1 in the time of gamma=9cm-1 and impression electric field being 7x107 V/m in the time of impression electric field being 5x107 V/m.

[0025] The response time by the 2 light-wave alligation of the photorefractive element produced in the [example 4] example 3 was measured. 2 light-wave alligation was performed on the same conditions as an example 3, and measured change of the probe

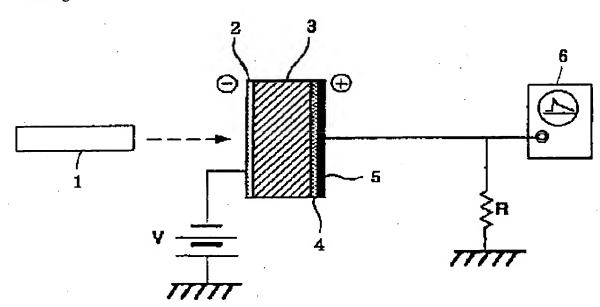
light reinforcement when turning on pump light. In order to evaluate the time amount of the standup of the probe light reinforcement at this time, as shown in $\frac{drawing 6}{dt}$, it is (1) type about measurement data. (1) y=A(1-exp(-x/tau))+B

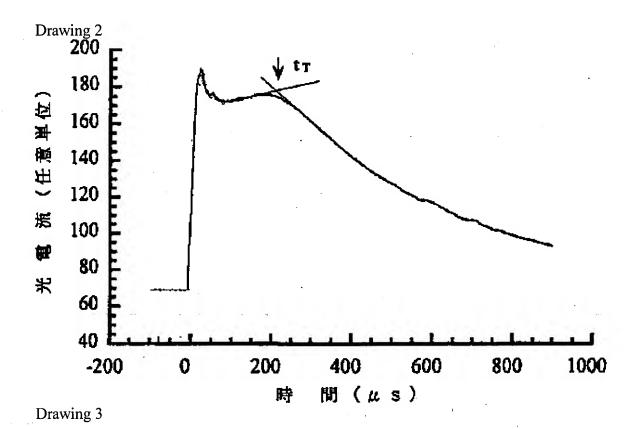
It was alike, with the least square method, and the response time tau was found. Consequently, the response time in case impression electric field are 5x107 V/m was 50msec(s).

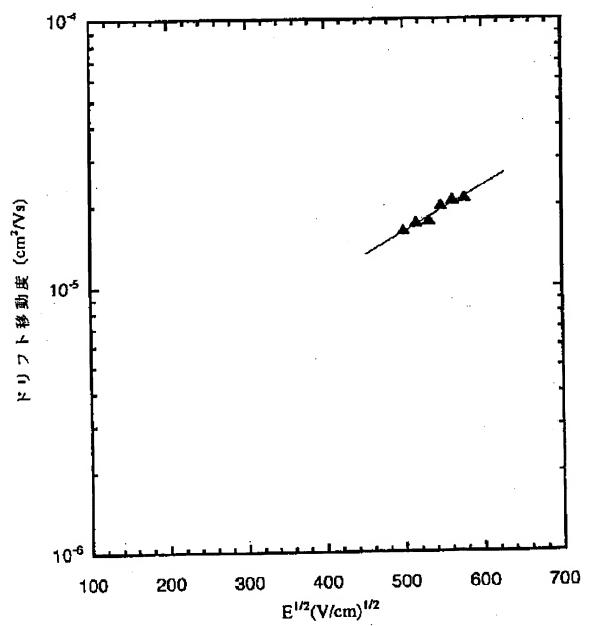
[0026] The response time by the 2 light-wave alligation of the photorefractive constituent which uses a polyvinyl carbazole as the base as a [example of comparison] hole transportability polymer was measured. The photorefractive element for measurement is the following, and was made and produced. First, the polyvinyl-carbazole 54.9 weight section, 4 - (N and N-diethylamino. -(beta)- the nitro styrene (DEANST) 14.9 weight section, the ethyl carbazole 30 weight section, and C600.2 weight section were dissolved in toluene / cyclohexanone (4:1) mixed solvent (0.1g/(ml)), and the sample solution was produced) Next, after carrying out the cast of the sample solution on the ITO electrode and being air-dry at a room temperature for 3 hours, the vacuum drying was performed at the room temperature for 12 hours. Then, the substrate was warmed at 60 degrees C, on both sides of the 100-micrometer Teflon spacer, lamination and weight were carried and two substrates were left at the room temperature for 12 hours. The sample with a thickness of 100 micrometers was obtained by this approach. When the response time by 2 light-wave alligation was measured by the same approach as an example 4 using this sample, the response times in case impression electric field are 5x107 V/m were 1.18sec(s) as shown in drawing 7. [0027]

[Effect of the Invention] By using the constituent of this invention, it has high-speed responsibility, the practical polymer system photorefractive ingredient which can be used for stability can be obtained, and it can be used for material for optical recording with quick drawing speed, the high-speed component ingredient for optical information processing, etc.

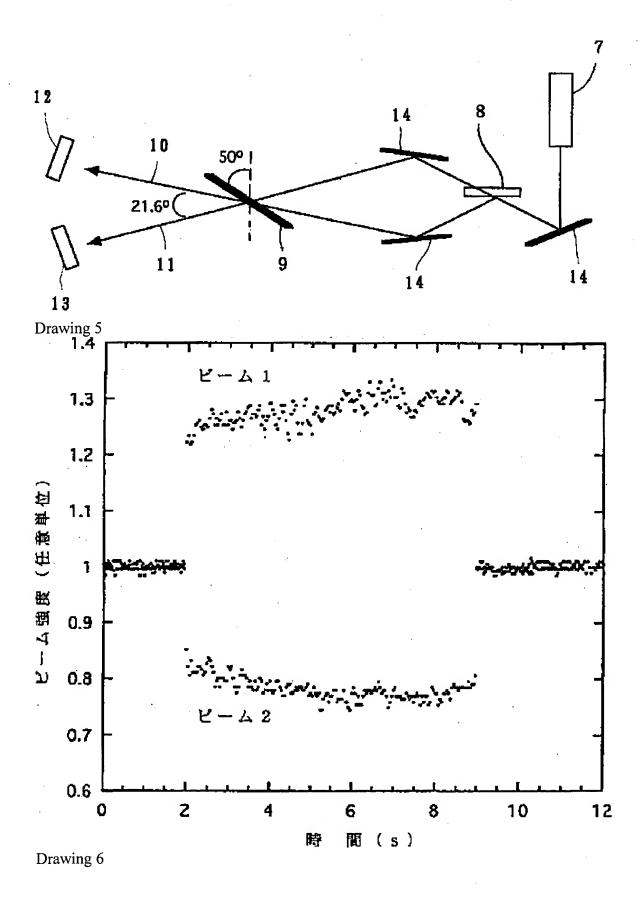
Drawing 1

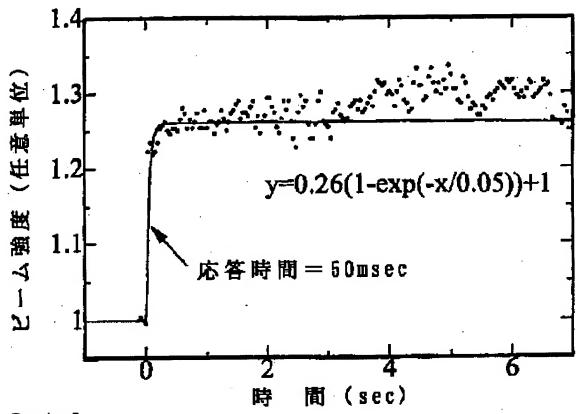




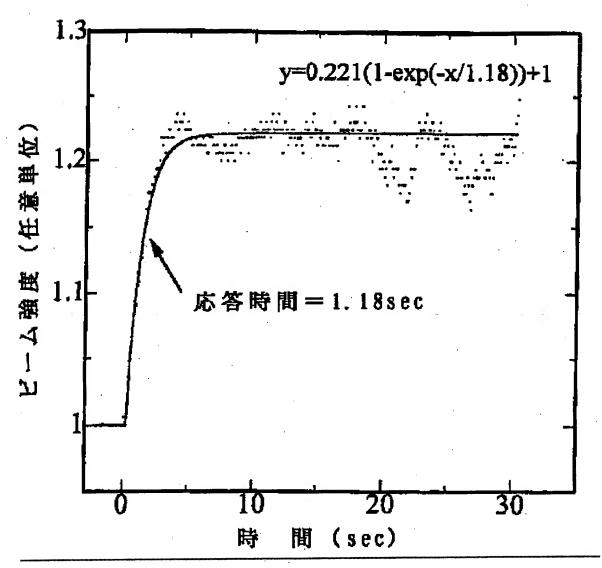


Drawing 4





Drawing 7



[Translation done.]